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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6 : C02F 1/469, B01D 61/44, 61/46

A1

(11) International Publication Number: WO 98/17590

(43) International Publication Date: 30 April 1998 (30.04.98)

(21) International Application Number: PCT/US97/19325

(22) International Filing Date: 22 October 1997 (22.10.97)

(30) Priority Data:
60/028,851 23 October 1996 (23.10.96) US

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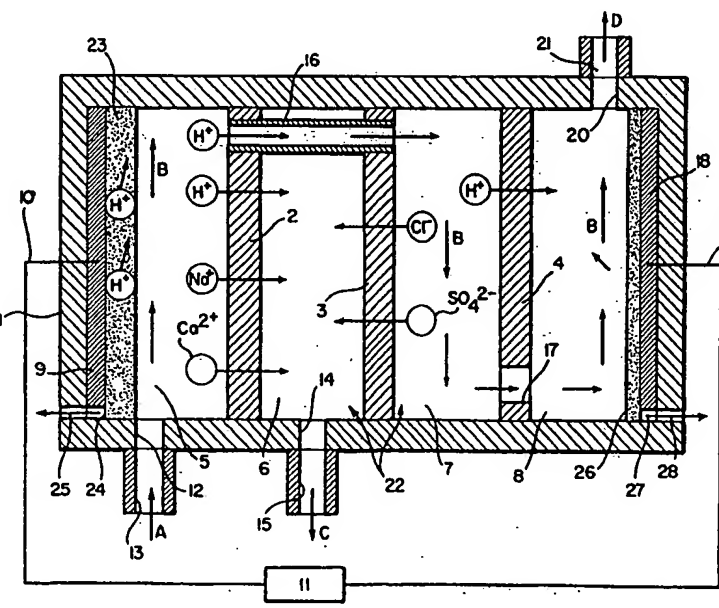
(81) Designated States: CN, SG, US, European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

Published
With international search report.

(54) Title: ELECTRODIALYZER AND PROCESS FOR DESALINATION



(57) Abstract

A multi-chamber electrodialysis system (1) is described which provides for initial electrolyte flow into an anodic chamber (5), across a succession of ion-exchange membranes (2, 3, 4), into at least one desalting chamber (5, 7), and exiting the cathodic chamber (8) with the desired final salt concentration. A concentration chamber (6) is provided which is isolated from the flow path by ion-exchange membranes (2, 3, 4). Salts and fluid are withdrawn from the concentration chamber (6), preventing insoluble salt formation on the ion-exchange membranes and allowing for continuous electrodialysis without reversing the polarity of the system. The system (1) provides the advantages of increased productivity due to the capacity for continuous electrodialytic operation and a simplified system structure.

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ELECTRODIALYZER AND PROCESS FOR DESALINATION

BACKGROUND

The present invention relates to an electrodialyzer unit for electrolyte separation using selective membranes under the action of an electromotive force treated in solution. In particular, this invention relates to a multi-chamber apparatus for desalting electrolyte aqueous solutions with ion-exchange membranes, and to a process for desalination.

Apparatus for desalting electrolyte aqueous solutions is known and in particular there are electrodialyzers which comprise a number of chambers formed in the housing of the apparatus by partitions of selective ion-exchange membranes through which solutions of salts are moved. In the end chambers there are electrodes connected to the direct electric current supply. Applied across the membranes, the electric current makes the chambers in turn enriched and poorer with electrolyte. This process results in separation of the initial solution of electrolyte into desalted and concentrated solutions.

As described in U.S. Patent No. 4,525,259, an electrodialyzer comprises a line of concentrating chambers between which are situated chambers with diluted solution. This line of chambers has a cathodic chamber with a cathode at one end and an anodic chamber with an anode at the other end. In the chambers adjacent to the chambers which contain electrodes, the diluted solution is maintained to prevent migration of ions from concentrated solutions in the concentration chambers into the chambers with the electrodes in them. All the chambers of this electrodialyzer have input and output pipe branches for corresponding aqueous solutions. Also, the apparatus has two tanks for catholyte and anolyte collection which are in

communication with corresponding electrode chambers. These tanks may be isolated from the electrode chambers during the process of solution recirculation between the chambers.

Another common electrodialyzer (F.N. Meller, "Electrodialysis - Electrodialysis Reversal Technology," IONICS, Incorporated, March 1984, pp. 53-56) comprises a housing inside of which are set alternately with cation- and anion-exchange membranes arranged parallel to each other to form a line of chambers; an anodic chamber with an anode set at one end wall and a cathodic chamber with a cathode at the other end wall with at least one pair of concentration/desalting chambers between them.

The anodic chamber is formed by the internal surfaces of the housing walls and by one side of the first cation-exchange membrane, the other side of which, together with the internal surfaces of the housing walls and one side of the anion-exchange membrane, forms a concentration chamber which is adjacent to the anodic chamber. Another side of the anion-exchange membrane together with the internal surface of the housing walls and the next cation-exchange membrane form a desalting chamber. The last cation-exchange membrane, one side of which limits the adjacent desalting chamber, forms the cathodic chamber by its other side and the internal surface of the housing wall. The rest of the pairs of concentration/desalting chambers are formed in the same way.

Each chamber of this known electrodialyzer has an input pipe branch for initial aqueous solution. What is more, anodic and cathodic chambers have input pipe branches for anolyte and catholyte respectively, while each concentration and desalting chamber has output pipe branches for concentrated and desalted solutions. This known electrodialyzer is equipped with a relay unit for electrode polarity reversal and with a device for

redirecting hydraulic flows from the desalting and concentration chambers.

The following describes the normal operation of the known electrodialyzer. An aqueous solution of electrolyte is fed simultaneously into all chambers through the respective pipe branches, then a direct electric current is supplied to the electrodes. The process of electrolysis takes place in anodic and cathodic chambers and results in anolyte and catholyte formation respectively. Under the influence of the direct current the ions of the salts diffuse through ion-exchange membranes into the concentration chamber realizing the desalting of aqueous solution in the desalting chamber and concentrate formation in the concentration chamber. Then the desalted solution and the concentrate are withdrawn out of the chambers via the respective pipe branches.

Electrolysis leads to a disturbance of the conditions of electric neutrality of aqueous solution at the interface between the ion-exchange membrane and the aqueous solution, and inevitably results in the formation of hardly soluble salts on the ion-exchange membranes. These salts reduce the permeability of the membranes. Owing to loss of conductivity, the desalting process stops.

To restore the desalting ability of the electrodialyzer it is then necessary to reverse the electrode polarity and reverse the directions of the hydraulic flow from the desalting and concentration chambers and thus change the direction of ion migration through the ion-exchange membranes. Electrochemical dissolution of sedimentary salts results in formation of an intermediate solution which is brought out of both chambers of each pair.

Then the first output pipe branches are shut while the second output pipe branches in the desalting and concentration chambers are opened such that the functions of these chambers are interchanged and the concentrate and the desalted solution are brought out of the chambers via the second pipe branches. When the desalting ability of the electrodialyzer is restored, the working cycle is repeated.

A common disadvantage of prior art devices such as that of the electrodialyzer which has been described herein, consists of low specific productivity caused by the inevitable formation of the sedimentary hardly soluble salts on the ion-exchange membranes and the resulting loss of conductivity necessitating a halt of the process of aqueous electrolyte solution desalting. The need to periodically restore the desalting apparatus, caused by the formation of the sedimentary hardly soluble salts on the membranes, requires additional means and measures to eliminate them.

Thus, the common design of the electrodialyzer is complicated due to the units for electrode polarity reversion and for turning hydraulic flow in directions opposite to their working ones. Besides, additional energy expenditures for electrochemical dissolution of these sedimentary salts are necessary. What is more, due to the need to feed the initial solution into each chamber and the need to form and withdraw intermediate- and end-products from each chamber, the usual design of the electrodialyzer is also complicated by the large number of pipe branches which result in inconvenience in assembly and maintenance and also leads to an increase in the total material required.

The closest analog of the electrodialyzer and of the electrolyte desalination process is described in

International Application No. PCT/GB93/00675 (WO93/20012) and U.S. Patent No. 5,589,050. However, both the apparatus and technology have a drawback associated with polarization and with discharge of gaseous products from the surface of electrodes, leading to an additional energy consumption due to increase in electric and hydraulic resistance caused by the synthesis and accumulation of air pockets inside electrode chambers.

The entire disclosures of International Application No. W093/20012 and U.S. Patent No. 5,589,050 are incorporated herein by reference.

SUMMARY

According to one aspect of the present invention, there is provided an electrodialyzer having a container with an input for supplying aqueous solution to be desalinated and an output for carrying substantially desalinated solution out of the container. At least one membrane is adapted to provide an anion exchange, and at least one membrane is adapted to provide a cation exchange. A non-linear continuous flow path is provided from the input to the output across the surfaces of the membranes, and a concentration chamber is separated from the flow path by, on one side an anion exchange membrane, and on the other side a cation exchange membrane. The electrodialyzer also has means for drawing off fluid accumulating in the concentration chamber.

Preferably, the electrodialyzer further has means for acidifying the aqueous solution near the beginning of the flow path, and means for neutralizing it towards the end of the flow path. The electrodialyzer may also have an anode at the beginning of the flow path in abutment with the container and covered with a cation-exchange membrane.

According to a second aspect of the present invention, there is provided a process for desalinating an aqueous solution. The process includes the steps of providing a housing having a succession of chambers including a concentration chamber bounded by a membrane adapted to provide anion exchange and a membrane adapted to provide cation exchange, supplying the aqueous solution to the succession of chambers in such a way that it flows in a continuous, non-linear flow path from an input to an output across the surfaces of the membranes, applying an electrical bias to the solution to cause ions to form and to migrate from the flow path into the concentration chamber, and drawing off fluid accumulating in the concentration chamber.

The concentration chamber is separated from the flow path by the anion exchange membrane and the cation exchange membrane. Preferably, the process further comprises causing the aqueous solution to be acidified at the beginning of the flow path and to be neutralized at the end of the flow path.

According to a third aspect of the present invention, there is provided an electrodialyzer comprising a housing with pipe branches for initial aqueous solution input and desalted water and concentrate output, a line of parallel chambers inside the housing being limited with the walls formed by the cation-exchange and anion-exchange membranes alternating with each other; an anodic chamber at one end wall of the housing, then at least one pair of concentration and desalting chambers, then a cathodic chamber at the other end wall of the housing. The anodic chamber with the anode in it is formed by the internal surface of the end wall of the housing and by a cation-exchange membrane, the other side of which limits the adjacent concentration chamber.

The opposite side of the concentration chamber is formed by an anion exchange membrane, while the opposite side of the membrane limits the desalting chamber, the opposite side of which is formed by a cation-exchange membrane. The opposite side of the cation-exchange membrane and the second end wall of the housing limit the cathodic chamber with a cathode disposed in it. The cathode and the anode are connected to the source of direct electric current. An advantage of the invention is that the pipe branch for initial aqueous solution input is disposed in the anodic chamber and the pipe branch for desalted water output is disposed in the cathodic chamber while the pipe branch for concentrate output is disposed in hydraulically isolated concentration chambers of each pair of chambers.

The internal volume of the anodic chamber successively communicates with the desalting chamber via hermetic channels provided in the concentration chambers of the pairs, while the last chamber in the line communicates with the cathodic chamber via an opening provided in the cation-exchange membrane. Furthermore, the anode in the chamber is fixed to the internal end wall of the housing and has an opening which is aligned with the opening in the end wall of the housing while an additional cation-exchange membrane is pressed to the surface of the anode from the internal side of the anode chamber.

One advantage of the present invention lies in the fact that its structure provides the necessary conditions for a continuous technological cycle of desalting of the initial electrolyte aqueous solution and thus increases the apparatus productive capacity with a reduction of energy consumption.

The structure of the anodic chamber, i.e. with the anode disposed at the end of the housing wall and with

direct contact between the anode surface and a cation-exchange membrane from the internal side of the chamber, this membrane being additionally provided in the anodic chamber, causes initiation of proton (H+) activation. At the boundary of contact of the anode surface and the damp cation-exchange membrane electrolysis takes place resulting in the formation of synthetic acid from water, which diffuses through the cation-exchange membrane into the anodic chamber and acidifies the initial aqueous solution fed into the chamber, thus providing the best desalting conditions. Aligned openings in the anode and in the end wall of the housing are preferably provided to form a channel via which gaseous products of decomposition are taken out of the chamber during electrolysis, ensuring good conditions for electrochemical synthesis in the anodic chamber.

Due to input of initial aqueous solution through the anodic chamber only and output of the final product from the cathodic chamber only as well as due to hydraulic isolation of the concentration chambers ensuring the flow of anolyte through these chambers, a continuous technological cycle takes place in the electrodialyzer and any hydraulic losses are substantially eliminated.

The design of the cathodic chamber, in which the cathode is located at the end of the housing wall and with direct contact with an anion-exchange membrane additionally introduced into the cathodic chamber, causes initiation of hydrogen hydroxyl (OH^-) activation. At the boundary of contact of the cathode surface and the moist anion-exchange membrane, the electrolysis takes place, leading to the formation of synthetic base from water, which diffuses through the anion-exchange membrane into the cathodic chamber and neutralizes the excess of acid fed from the desalination chamber. This contact also prevents gases from entering the cathodic chamber, thus ensuring the best

conditions for desalination in the apparatus. Aligned openings in the cathode and in the end wall of the housing provide a channel via which gaseous decomposition products are removed from the chamber during electrolysis, maintaining a stable current density at the cathode, thus increasing the capacity of the apparatus.

Also the structure of the electrodialyzer of the invention provides technological conditions which prevent the formation and deposition of hardly soluble sediments on the ion-exchange membranes thus excluding the necessity of periodically stopping the apparatus for electrochemical dissolution of sediments and hence increasing the productive capacity of the apparatus. At the same time, energy consumption is reduced and the equipment of the apparatus is simplified; instead of a minimum of ten input/output pipe branches in the previous art apparatus, there can be a minimum of three such branches in the apparatus according to the invention (attached to one pair of chambers between the electrode chambers).

Other objects and features of the present invention will be apparent from the following detailed description and drawings which illustrate preferred embodiments of the invention.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic view of an electrodialyzer including one concentration chamber and one desalting chamber in longitudinal section according to the invention; and

FIG. 2 is a general view of an electrodialyzer according to the invention with three pairs of chambers in longitudinal section.

DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

Referring now to the drawings, where like elements are designated by like reference numerals, there is shown in FIG. 1 an electrodialyzer constructed in accordance with the present invention. The electrodialyzer has a hermetic housing 1 (FIG. 1) inside of which are mounted in parallel, alternately a cation-exchange membrane 2, an anion-exchange membrane 3, and a cation-exchange membrane 4.

The membranes 2, 3, 4 divide the cavity of the housing 1 into an anodic chamber 5, a concentration chamber 6, a desalting chamber 7 and a cathodic chamber 8. The chambers 5, 6, 7, 8 within the housing 1 extend parallel to each other. The electrode chambers 5 and 8 are situated at the end walls of the housing 1.

The anodic chamber 5 is formed by the inner surfaces of the end and side walls of the housing 1 and by one side of the cation-exchange membrane 2. On the inner surface of the end wall of the housing 1 there is fixed an electrode which is the anode 9 provided with a lead 10 which extends outwardly from the housing 1 and is connected to a positive terminal of a direct current power supply 11. The bottom wall of the anodic chamber 5 has an opening 12 with a pipe connection 13 for input of initial aqueous electrolyte solution.

The concentration chamber 6 is formed by the inner surfaces of the side walls of the housing 1, the other side of the cation-exchange membrane 2 and one side of the anion-exchange membrane 3. The bottom wall of the chamber 6 has an opening 14 with a pipe connection 15 for output of the concentrate. In the upper parts of the cation-exchange membrane 2 and anion-exchange membrane 3 there is a pipe 16. The pipe 16 represents a channel connecting the anode

chamber 5 and the desalting chamber 7 and serves for anolyte flow.

The desalting chamber 7 is formed by the inner surfaces of the side walls of the housing 1, the other side of the anion-exchange membrane 3 and one side of the cation-exchange membrane 4. In the lower part of the membrane 4 there is an opening 17 for communication of the desalting chamber 7 with the cathodic chamber 8. The concentration chamber 6 and the desalting chamber 7 make one pair (or block) 22 of chambers. Depending on the demands of initial aqueous solution filtration there may be several pairs (blocks) of chambers in the electrodialyzer.

The cathodic chamber 8 is formed by another side of the cation-exchange membrane 4, the inner surfaces of the side walls of the housing 1 and its end wall on which the electrode (cathode 18) is fixed. The lead 19 of the cathode 18 extends outwardly and is connected to a negative terminal of the power supply 11. In the cathodic chamber 8 an opening 20 is provided into which a pipe connection 21 is inserted for output of the desalted solution.

To intensify the process, an additional cation-exchange membrane 23 is applied to anode 9. In the anode 9, an opening 24 is provided which is aligned with an opening 25 in the end wall of the housing 1. The openings 24 and 25 form a channel for withdrawal of the gaseous products of the acid synthesis from the anodic chamber. The process is further enhanced by using an anion-exchange membrane 26 applied against the cathode 18.

An opening 27 is provided in the cathode 18. The opening 27 is aligned with the opening 28 in the end wall of the housing. The openings 27 and 28 form a channel for removing gaseous products of the basic synthesis from the cathodic chamber 8.

Aqueous solution is fed in the direction marked with the arrow A. Arrows B designate the directions of the aqueous electrolyte solution flow inside the housing 1 of the electrodialyzer. Concentrate and desalted aqueous solution/final product are withdrawn in the directions designated by arrows C and D, respectively.

In FIG. 1 are also designated:

H^+ - proton
Na^+ - sodium cation
Ca^{2+} - calcium cation
Cl^- - chlorine anion
SO_4^{2-} - sulphate anion

The arrow with the circle at its end in which the charge of the ion is indicated shows the direction of ion movement.

FIG. 2 shows an electrodialyzer having three pairs 22 of concentration/desalting chambers 6, 7. In the first pair 22 of chambers the pipe 16 is provided in the upper part of the chamber 6, in the second pair 22 of chambers the pipe 16 is provided in the lower part of the chamber 6 and in the third pair 22 of chambers the pipe 16 is provided in the upper part of the chamber 6. The above mentioned arrangement of the pipes 16 ensures continuous and uniform flow of aqueous solution through all chambers 5, 6, 7, 8.

The electrodialyzer of FIGS. 1 and 2 operate as follows. Desalting of aqueous solution takes place in the process of continuous filtration when passing aqueous solution through the anodic chamber 5 (FIGS. 1, 2), the desalting chamber 7 and the cathodic chamber 8. Initial aqueous solution, salt water for instance, is fed to the anodic chamber 5 through the input pipe connection 13. At

the same time, electric current is supplied from the power supply 11 to the anode 9 and to the cathode 18 with density which provides pH=2 in the concentration chamber 6. This pH value prevents precipitation of hardly soluble salts onto the membranes 2, 3.

Under the action of electric current in the region where the surface of the anode 9 contacts the damp cation-exchange membrane 23, electrolysis occurs and synthetic acid (shown in FIG. 1 as protons H^+) is formed. The synthetic acid diffuses through the cation-exchange membrane 23 into the anodic chamber 5 and acidifies the initial aqueous solution (fed via the pipe connection 13), while the gaseous product of decomposition is taken off through the openings 24 and 25 outwardly to a gas-holder (not illustrated).

Then acid aqueous solution is fed through the pipe 16 to the desalting chamber 7 where, under the action of direct electric current, salt ions (Cl^-, SO_4^{2-}) from aqueous solution diffuse through the membrane 3 to the concentration chamber 6, while Ca^{2+} and Na^+ ions diffuse through the membrane 2 from the anodic chamber 5 to the concentration chamber 6. The gaseous product is taken away from the anodic chamber 5 via the openings 24 and 25.

Acid diffuses through the membrane 4 from the desalting chamber 7 to the cathodic chamber 8. Thus desalting of aqueous solution with the concentrate formation takes place in the chamber 6, from which the concentrate is withdrawn via a pipe connection 15 in the direction designated by arrow C. Acid aqueous solution is fed through the opening 17 in the membrane 4 to the cathodic chamber 8 where, under the action of electric current, the electrolysis of aqueous solution takes place with formation of hydroxyl ions (OH^-) in a quantity which is equivalent for neutralization of the acid fed to the

cathodic chamber 8. Then the desalted aqueous solution is withdrawn through the pipe connection 21 from the cathodic chamber 8. The gaseous product is taken away from the cathodic chamber 8 via the openings 27 and 28.

FIG. 2 shows an electrodialyzer having three pairs 22 of chambers 6 and 7 between the electrode chambers 5 and 8. The electrodialyzer of FIG. 2 otherwise works in the same way as the electrodialyzer of FIG. 1. The initial solution of salt water, for instance, is fed through the input pipe connection 13 of the anodic chamber 5. Anions (Cl^-, SO_4^{2-}) diffuse through the anion-exchange membrane 3 from the desalting chamber 7 of the second pair 22 of chambers to the concentration chamber 6 of the same pair 22. Thus desalting of aqueous solution takes place in the desalting chamber 7 of all pairs 22 and the concentrate is formed in all concentration chambers 6. The concentrate is removed from all concentration chambers 6 through the output pipes 15. Desalted aqueous solution is removed from the cathodic chamber 8 through the pipe 21.

Thus, the illustrated electrodialyzers provide the conditions for a continuous cycle of aqueous solution desalting with maximum possibility of electrochemical interaction of aqueous solution with the structural elements of the apparatus in its working chambers, thus increasing the productive capacity of the apparatus with simultaneous reduction of total energy consumption.

The invention is applicable to the purification of aqueous solutions with excessive soluble salt impurities to achieve a required final concentration of salts. Also, the invention may be widely applied for the purification of water in the chemical, power, radioelectronic and medical industries as well as in municipal water supply systems.

The entire disclosure of U.S. Provisional Patent Application No. 60/028,851, filed October 23, 1996 is incorporated herein by reference.

The above description and drawings illustrate preferred embodiments which achieve the objects, features and advantages of the present invention. It is not intended that the present invention be limited to the illustrated embodiments. Any modification of the present invention which comes within the spirit and scope of the following claims should be considered part of the present invention.

What is claimed is:

1. An electrodialyzer, comprising:

an input for receiving an aqueous solution;

a concentration chamber for receiving concentrate from the aqueous solution, said concentration chamber being separated from said input;

a cathodic chamber in fluid communication with said input;

a cathode for applying current to the aqueous solution, said cathode being located in said cathodic chamber;

an anion exchange membrane located adjacent said cathode, said anion exchange membrane being located in said cathodic chamber; and

an opening for removing gaseous products from said cathodic chamber.

2. The electrodialyzer of claim 1, further comprising an anode for applying current to the aqueous solution, a cation exchange membrane located adjacent to said anode, and a second opening for removing gaseous products.

3. The electrodialyzer of claim 2, wherein said anode is located adjacent to said input.

4. The electrodialyzer of claim 3, further comprising a continuous and non-linear flow path connected to said input, said flow path including said cathodic chamber.

5. An electrodialyzer, comprising:

an input for receiving aqueous solution to be desalinated;

an anode and a cation exchange membrane contacting said anode;

a cathode and an anion exchange membrane contacting said cathode;

parallel chambers in fluid communication with said input, said chambers including an anodic chamber, a

desalting chamber, and a cathodic chamber, and wherein said input, said anode and said cation exchange membrane are located in said anodic chamber, and wherein said cathode and said anion exchange membrane are located in said cathodic chamber; and

outlets for gases generated in said anodic and cathodic chambers.

6. The electrodialyzer of claim 5, further comprising means for applying an electrical bias between the anode and the cathode such that the aqueous solution is exposed to the electrical bias substantially throughout said parallel chambers.

7. A process for desalinating an aqueous solution, said process comprising the steps of:

providing a housing having successive chambers defining a flow path, and a concentration chamber separated from the flow path and bounded by anion and cation exchange membranes;

supplying the aqueous solution to the successive chambers such that the aqueous solution flows from an input to an output successively across the surfaces of the membranes;

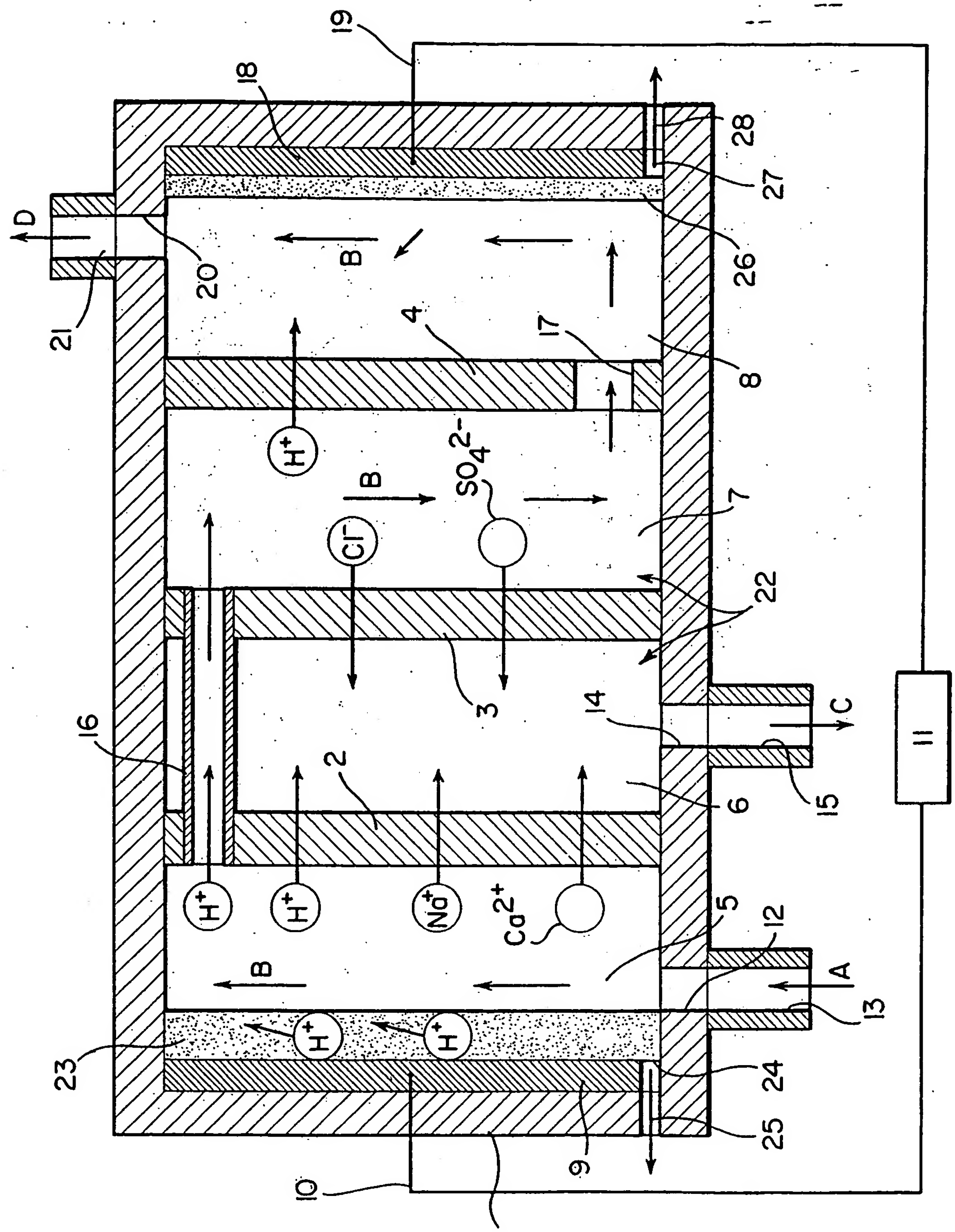
applying an electrical bias to the aqueous solution to cause ions to form and to migrate into the concentration chamber; and

drawing off fluid accumulated in the concentration chamber.

8. The process of claim 7, wherein the aqueous solution is acidified at the beginning of the flow path and neutralized at the end of the flow path.

9. The process of claim 8, wherein the flow path is non-linear.

10. The process of claim 9, further comprising the step of supplying the aqueous solution along the flow path from the input into an anodic chamber, across the cation exchange membrane, through a channel through the concentration chamber, into a desalting chamber, across the anion exchange membrane, and then through an opening in a second cation exchange membrane separating the desalting chamber from a cathodic chamber.



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Ca^{2+}
Cl^-
SO_4^{2-}

FIG. 1

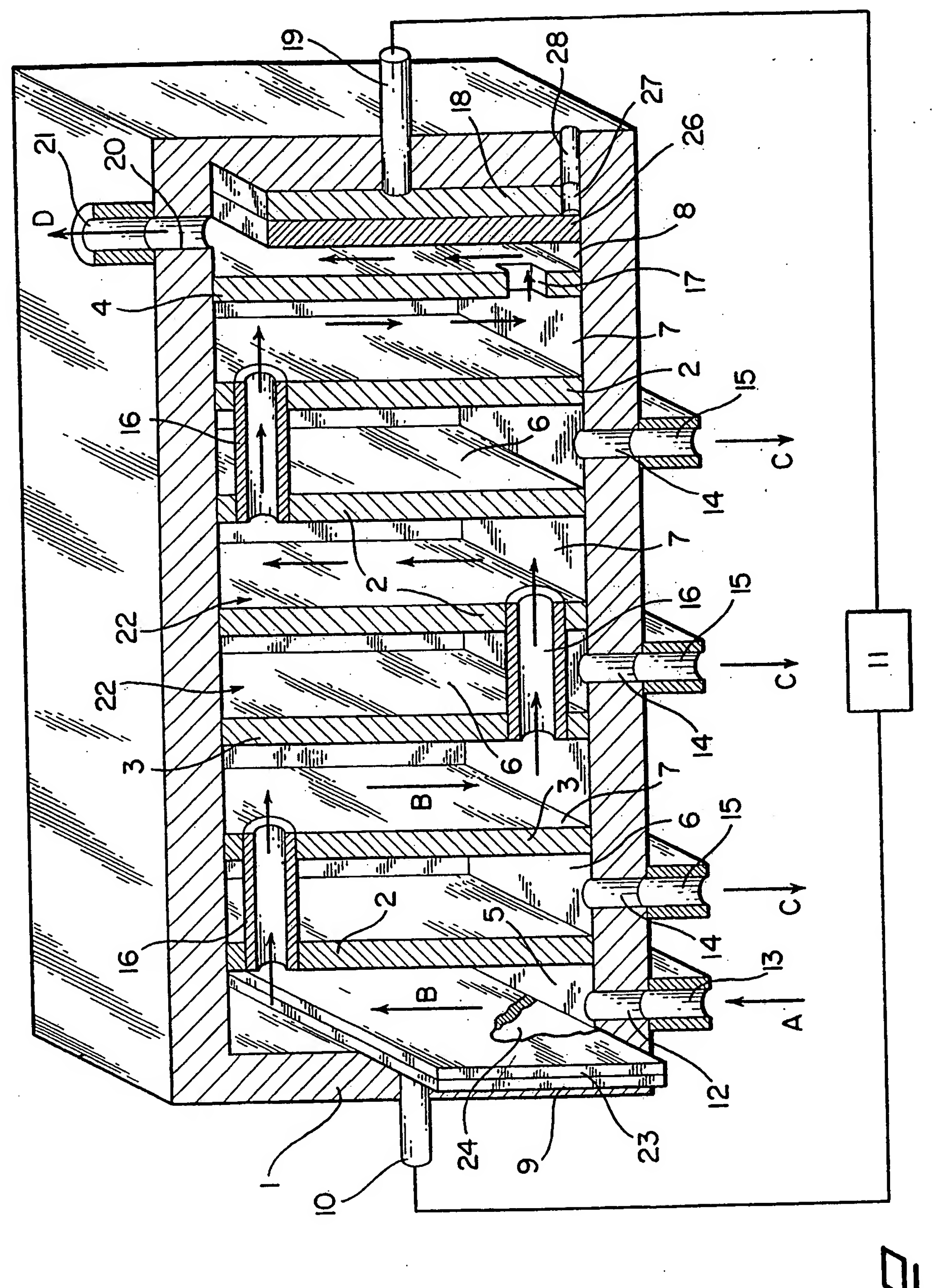

FIG. 2

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US97/19325

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :C02F 1/469; B01D 61/44, 61/46
US CL :205/748; 204/525, 529, 634, 635, 636
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 205/748; 204/525, 529, 634, 635, 636

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

none

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

none

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A,P	US 5,589,050 A (MEDVEDEV ET AL) 31 December 1996 (31-12-96), see claims 1-5.	1-10
A	WO 91/04782 A (MEDVEDEV ET AL) 18 April 1991 (18-04-91), see abstract and figures 1-2.	1-10

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

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